Signature of Cooper pairs in the non-superconducting phases of amorphous superconducting tantalum films

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Abstract

We have studied the magnetic field or disorder induced insulating and metallic phases in amorphous Ta superconducting thin films. The evolution of the nonlinear transport in the insulating phase exhibits a non-monotonic behavior as the magnetic field is increased. We suggest that this observation could be an evidence of the presence of localized Cooper pairs in the insulating phase. As the metallic phase intervenes the superconducting and insulating states in Ta films, this result further reveals that Cooper pairs also exist in the metallic ground state.

Superconductivity in homogeneously disordered two dimensional (2D) system is of particular interest because 2D is the lower critical dimension for both superconductivity and localization. Conventional theory predicts that in 2D the disorder- or magnetic field (B)-induced suppression of superconductivity leads to a direct superconductor-insulator transition (SIT) [1–5] in the zero temperature ($T \to 0$) limit. The "dirty boson" model [1, 2], which describes the superconducting phase as a condensate of Cooper pairs with localized vortices and the insulating phase as a condensate of vortices with localized Cooper pairs, assumes the presence of Cooper pairs on both sides of the SIT. The existence of localized Cooper pairs in the insulating phase of granular films is undoubted, however, whether Cooper pairs are present in the insulating phase of amorphous films is a more complicated issue [6]. Experimental evidence on the crossover from Bose insulator with nonzero pairing to Fermi electronic insulator without pairing in amorphous InO films [7], and the emergence of a magnetoresistance (MR) peak in several 2D amorphous materials [7–14], such as InO [7–11], MoSi [12, 13], and Bi [14], and the observation of activated resistances and magnetoresistance oscillations dictated by the superconducting flux quantum in patterned amorphous Bi films [15], suggest that Cooper pairs might be present in the insulating phase of homogeneously disordered superconducting films. More recently, the direct evidence for the existence of preformed Cooper pairs in non-superconducting states of amorphous InO [16], TiN [17], and NbN [18] films is revealed by scanning tunneling spectroscopy. However, question concerning whether the persistence of Cooper pairs in insulating phase is a generic or a material-based property of amorphous superconducting films remains.

While the nature of the insulating state is still an issue to be solved, the subject has attracted more attention because of the observation of the metallic phase in amorphous MoGe [19, 20] and Ta [21–24] thin films under weak magnetic fields. The unexpected metallic behavior, which intervenes the B- or disorder- driven SIT, is characterized by a drop in resistance (ρ) followed by a saturation to a finite value as $T \to 0$. This low field- or disorder- driven metallic phase is significantly different from the high field "quantum metal" observed in insulating Be films [25] or superconducting InO films [9] and TiN films [26], which is induced by high B fields on films that already exhibit insulating phase. Several theoretical models have been proposed to account for the emergence of the metallic ground state [27–31], including the quantum phase glass model [27, 28], the quantum vortex picture [29], and the percolation paradigm [30, 31]. However, a consensus on the mechanism for the

metallic behavior hasn't been reached yet.

In this work, we study the evolution of nonlinear current-voltage (I-V) characteristics in the insulating phase of amorphous superconducting Ta films with the increase of B field. The non-monotonic B field dependence of the dV/dI peak, which has been observed in all the films we have studied, suggests the existence of the localized Cooper pairs in both B- and disorder- driven insulating phases. Our study also implies that the nonlinear transport characteristics might be a consistent and sensitive probe to detect localized Cooper pairs.

Our Ta thin films are dc sputter deposited on Si substrate and are patterned into a 1mm wide and 5mm long Hall bar for the standard four point measurement using a shadow mask. The thickness of the films is between 6 nm and 2 nm. As reported before [21], the x-ray diffraction pattern of such films does not show any sign of local atomic correlation, so they are structurally amorphous and homogeneously disordered, as expected from the excellent wetting property of Ta on almost all substrates. The temperature dependence of resistivity measurement indicates that the superconducting transition temperature decreases continuously toward 0 K with increasing disorder and the transition exhibits no reentrant behavior, which further confirms the amorphous nature of these Ta films [6]. The magnetoresistance was measured by a lock-in with 1 nA ac current. The dV/dI trace was measured by a homemade ac+dc currents summing circuit which used a lock-in to modulate the dc bias current with a small ac amplitude at low frequency.

Recent studies on Ta films have shown that each phase displays remarkably different nonlinear I-V characteristics [21], offering an alternative criterion to identify phases which is fully consistent with that based on the T dependence of ρ. The superconducting phase is unique in exhibiting hysteresis in the I-V curve, which has been demonstrated to arise from a nonthermal origin [22]. As the system is driven into the metallic phase, the hysteresis evolves into the point of the largest slope in the continuous and reversible I-V implying that the nonlinear transport in the metallic phase ($d^2V/dI^2 > 0$) is also intrinsic and uncorrelated with electron heating effect [22]. The I-V characteristics and accompanying long electronic relaxation time in the superconducting and metallic phases can be well explained by the vortex dynamics picture [24]. The insulating phase is characterized by a peak structure in the dV/dI vs. I trace ($d^2V/dI^2 < 0$), and has been employed as a phenomenological symbol to identify the phase [24].

Qualitatively similar dV/dI peaks have been observed in the insulating phases of TiN

TABLE I. List of sample parameters: nominal film thickness t, normal state resistivity ρ_n at 4.2 K, and the observed phases at low temperature (S for the superconducting phase, M for the metallic phase, and I for the insulating phase). For samples exhibiting the superconducting phase, we list mean field T_c at B = 0, the critical magnetic field H_c as defined by the field at which the low temperature (60 mK for Ta 1 - Ta 3, 50 mK for Ta 4) resistance reaches 90% of the high field saturation value, and the superconducting coherence length calculated from $\xi = \sqrt{\Phi_0/2\pi B_c}$, where Φ_0 is the flux quantum.

Films	Batch	t(nm)	ρ_n(k$\Omega/\square$)	phase	T_c(K)	H_c(T)	ξ(nm)
Ta 1	1	5.6	1.42	S,M,I	0.65	0.82	20
Ta 2	1	4.6	1.85	S,M,I	0.54	0.68	22
Ta 3	1	5.1	2.16	S,M,I	0.38	0.58	24
Ta 4	2	4.1	2.28	S,M,I	0.26	0.33	32
Ta 5	3	2.8	4.62	M,I			
Ta 6	4	2.5	6.24	I			
Ta 7	4	2.5	8.00	I			
Ta 8	3	2.36	8.78	I			

[32], InO [33], and MoGe [34]. However, the origin of the insulating nonlinear transport might be different for various systems. The nonlinear I-V in strong insulators, especially the giant jumps of current at finite voltages in InO and TiN [32, 35], is believed to be a consequence of overheating of electrons [36]. We note that the amorphous Ta films we studied are significantly different from those systems. Firstly, the I-V characteristic in the insulating phase of Ta is continuous showing no sign of a bistability. Secondly, the validity of the proposed electrons overheating model [36] requires a steep temperature dependence of the resistance, which is the case for InO and TiN but not the case for Ta [24]. Furthermore, the non-monotonic B field dependence of the dV/dI peak structure in the insulating phase of Ta, especially the fact that the most pronounced non-monotonic feature appears in the B-induced insulating phase of low disordered films which have lower resistance and thus less likely to be overheated compared with the highly disordered films, as presented in this paper, indicates that the electron heating effect, if plays a role, could not be the only source for the nonlinear transport in the insulating phase of Ta films.

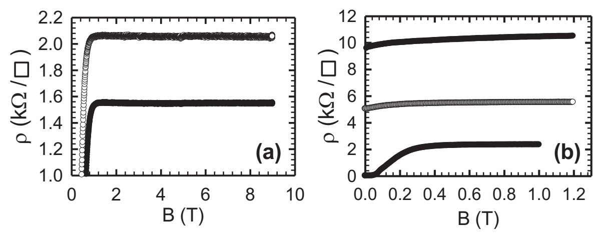

FIG. 1. (a) Magnetoresistance for Ta 1 (bottom trace) and Ta 2 (top trace) at T = 60 mK. (b) Magnetoresistance for (from bottom to top) Ta 4, Ta 5, and Ta 7 at T = 50 mK.

Conventionally, the B-induced insulating phase was studied by MR measurements [7–14]. The negative MR is often interpreted as a consequence of a conduction enhancement due to delocalization and/or breaking of the localized Cooper pairs which are believed to be present in the insulating phase [1, 2]. The positive MR is usually attributed to the response of unpaired electrons [12–14, 37, 38]. Figure 1 shows the MR measured on five Ta films with different degrees of disorder. Parameters of the films are summarized in Table I. The resistivity of each film increases monotonically with the magnetic field and eventually saturates, implying that the localized Cooper pairs in the insulating phase of Ta films, if present, are not detectable in the linear transport regime.

Figure 2(a) illustrates the evolution of differential I-V in the B-induced metallic and insulating phases in superconducting film Ta 1. For B $\leq$ 0.8 T (B $\geq$ 0.9 T), the dV/dI is a monotonically increasing (decreasing) function of the bias current, which characterizes the metallic (insulating) phase. At B = 0.85 T (thick solid line), the sign of d^2V/dI^2 is positive (negative) at high (low) bias currents as in the metallic (insulating) phase at lower (higher) fields. The non-monotonic dV/dI with respect to bias current was interpreted as an evidence that the insulating state near B_c consists of metallic domains connected by point contacts [23]. Fig. 2(b) shows the MR measured in the low current limit (1 nA) at three different temperatures within the low T regime. The presence of a crossing point at B_c = 0.83 T, which is defined as the critical field for metal-insulator transition, confirms that the transport at low currents at B = 0.85 T is insulating.

In this work, we focus on the evolution of dV/dI peak structure in the insulating phase with the increase of magnetic field. The height of the dV/dI peak in Fig. 2(a) initially increases with increasing B, reaching a maximum at B = 1.1 T, and then decreases with further increase of B. We use the normalized peak height h(B)/h(B=9T) as a quantitative

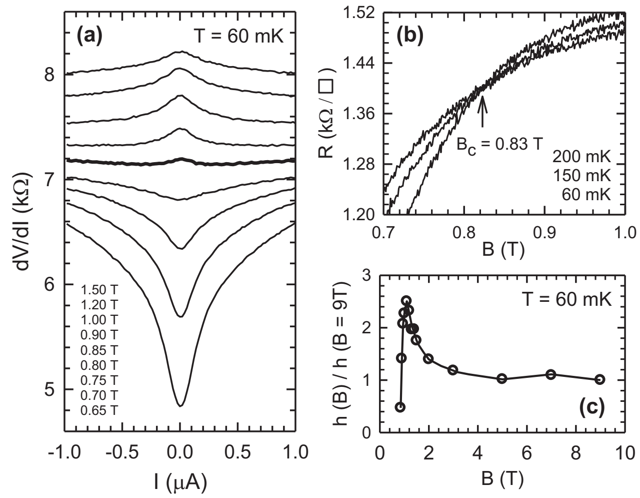

FIG. 2. (a) Evolution of dV/dI vs. I with increasing B for Ta 1 at T = 60 mK. Each trace is vertically shifted successively for clarity. (b) Magnetoresistance for Ta 1 at the indicated temperatures (from top to bottom below the crossing point). The crossing point at 0.83 T marked by an arrow defines the critical field B_c for metal-insulator transition. (c) Normalized height of the dV/dI peak structure h(B)/h(B=9T) vs. B for Ta 1 at T = 60 mK. Symbols are experimental data and solid lines are to guide the eye.

measure of the non-monotonic feature we observe and plot it as a funtion of the magnetic field in Fig. 2(c). h(B)/h(B=9T) increases with B field until reaching a prominent maximum and then decreases, showing a general trend of saturation. In the context of “dirty boson” model, the dV/dI peak is attributed to the current-induced delocalization of the localized Cooper pairs [21]. We thus expect that the dV/dI peak height initially grows as the system is driven into the insulating phase where Cooper pairs are localized. Above a certain magnetic field ($\sim$ 1.1 T for Ta 1), however, the population of the localized Cooper pairs would decrease with increasing B because of the B-induced pair breaking mechanism. As the population of the localized Cooper pairs decreases, the effect of their current-induced delocalization would consequently decrease, resulting in a reduced dV/dI peak height. We suggest that this non-monotonic feature is a result of superconducting correlations. Although the electron heating effect and/or Coulomb interaction between normal electrons might also lead to dV/dI peak structures, neither of them could be responsible for the observed non-monotonic evolution of the peak structure as the magnetic field is increased. This result is thus interpreted as a signature of the localized Cooper pairs in the B-induced insulating phase of superconducting

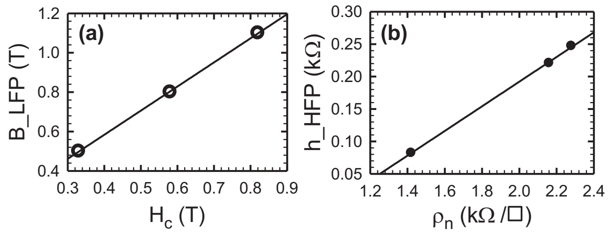

FIG. 3. (a) The magnetic field where the low field maximum occurs in the h(B)/h(B=9T) vs. B trace as a function of the critical magnetic field H_c for 3 low disordered films, Ta 1, Ta 3, and Ta 4. (b) The height of the dV/dI peak where the high field maximum takes place as a function of the normal state resistivity ρ_n for Ta 1, Ta 3, and Ta 4. Symbols are experimental data and solid lines are linear fits.

Ta films.

Such non-monotonic feature in the dV/dI peak height vs. B plot is evident for all low disordered Ta films we have studied. Figure 3(a) shows the B field where the maximum occurs as a function of the critical magnetic field H_c for Ta 1, Ta 3, and Ta 4. The data falls into a line in an almost perfect fashion. As H_c is directly related to the superconducting coherence length ξ, this result further suggests the intrinsic link between the non-monotonic feature and superconducting correlations. In addition to the prominent maximum in the dV/dI peak height vs. B plot at $\sim$ 1 T or below, a shallow peak at $\sim$ 7 T, as shown in Fig. 2(c) is also observed in the B-induced insulating phase of all superconducting films. The origin for this high field feature, which is most likely not due to superconductivity related mechanism [39, 40], is not clear at present and might require systematical studies at higher magnetic fields. To distinguish the low field non-monotonic feature from the high field maximum, we name the former as low field peak (LFP) and the latter as high field peak (HFP). The height of the HFP is found to show a linear dependence on the normal state resistivity, as shown in Fig. 3(b).

In addition to low disordered samples which exhibit superconducting behaviors at low T and low B, we also studied the evolution of dV/dI as a function of B field for highly disordered films that are insulating at low T at B = 0. Figure 4 shows dV/dI spectra and normalized peak height vs. B for two insulating samples. A low field maximum of the dV/dI peak height, which is less pronounced than that in Fig. 2, emerges at $\sim$ 1 T as shown in Fig. 4(a) and 4(b) for Ta 7. Figure 4(c) shows the dV/dI spectra of Ta 8, a sample with

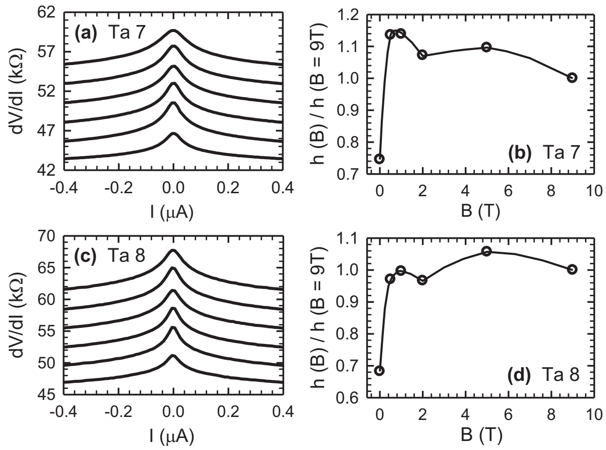

FIG. 4. (a) Evolution of dV/dI vs. I for Ta 7 at T = 60 mK at B = 0 T, 0.5 T, 1 T, 2 T, 5 T, and 9 T. Each trace is vertically shifted successively. (b) Normalized height h(B)/h(B=9T) vs. B for Ta 7 at T = 60 mK. Solid lines are to guide the eye. (c) Evolution of dV/dI traces (vertically shifted) for Ta 8 at the same temperature and magnetic fields as in panel (a). Ta 8 is more disordered than Ta 7 by the measure of ρ_n. (d) Normalized height as a function of B field for Ta 8 at T = 60 mK. Solid lines are to guide the eye.

a higher degree of disorder than Ta 7. As indicated in Fig. 4(d), a fairly weak maximum appears at $\sim$ 1 T in the dV/dI peak height vs. B plot. The weakening of the B-induced Cooper pairs breaking effect with the increase of disorder, could be a consequence of a reduced population of Cooper pairs as the degree of disorder is increased. Alternatively, it might be caused by smearing of the B-induced Cooper pairs breaking effect by disorder [40], or by electron heating which, if contributes to the dV/dI peak structures, would play an increasingly more important role with the increase of disorder.

A high field maximum is also observed in these highly disordered insulating films, as shown in Fig. 4(b) and 4(d), which probably has the same origin as the HFP for low disordered samples. The heigths of the dV/dI peaks at 1 T (open symbols) and 5 T (filled symbols), and their ratio are plotted as a function of ρ_n in Fig. 5, for 3 highly disordered films, Ta 6 - Ta 8. The peak heights grow with the increase of disorder, and a linear dependence on ρ_n is observed for the peak at B = 1 T. The ratio of the peak heights at 1 T and 5 T decreases from above 1 to below 1, as ρ_n is increased, indicating that the LFP is more sensitive to the disorder.

The evolution of dV/dI peak structure with the increase of B field for films with various

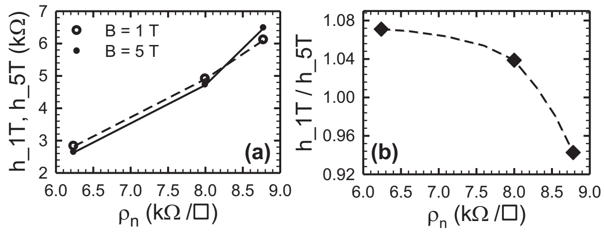

FIG. 5. (a) dV/dI peak height at B = 1 T (open symbols) and 5 T (filled symbols) vs. normal state resistivity ρ_n for 3 highly disordered films, Ta 6 - Ta 8. (b) Ratio of the dV/dI peak heights at B = 1 T and 5 T as a function of ρ_n for Ta 6 - Ta 8. Solid and dashed lines are to guide the eye.

degrees of disorder, as shown in Fig. 2 - Fig. 5, suggests that Cooper pairs exist in both B- and disorder- induced insulating phases of Ta films and can be detected by nonlinear electronic transport. Although this result could not exclude the possible contributions of unpaired normal electrons and/or electron heating to the dV/dI peak in the insulating phase, neither of them could produce the observed low-field non-monotonic feature, which would not be possible without the presence of Cooper pairs. In a recent paper [24], we mapped the phase diagram of Ta thin films in B-T-disorder space, which indicates that the superconducting phase is completely surrounded by the metallic phase prohibiting a direct SIT at any disorder. Thus our result further reveals the presence of Cooper pairs in the metallic phase which intervenes the superconducting and insulating regimes, as proposed by Phillips *et al* [27, 28].

Although a lot of theoretical efforts have been devoted to understand the transport properties of disordered 2D superconductors in the linear current response regime, not much theoretical work has been carried out to study the nonlinear I-V. One of the major reasons might be that very limited experimental results in the nonlinear current response regime have been reported. The work presented here would stimulate the development of more comprehensive theoretical models to account for the transport behaviors in the nonlinear I-V regime of disordered superconducting films.

To summarize, we have reported that in the insulating transport regime of homogeneously disordered Ta films, the height of the dV/dI peak structure experiences a non-monotonic change with the increase of magnetic field. This observation suggests that Cooper pairs persist into the insulating phase of the films, and further implies the presence of Cooper

pairs in the metallic phase which intervenes both the B-induced and the disorder -induced superconductor-insulator transition. Our work indicates that, compared with the traditional magnetoresistance measurement, nonlinear transport characteristics might be a more sensitive probe of localized Cooper pairs.

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